

**Supporting information**

**Achieving Balanced Intermixed and Pure Crystalline Phases in  
PDI-based Non-fullerene Organic Solar Cells via Selective  
Solvent Additives**

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**Table captions**

**Table S1.** Physical properties and solubility parameters calculated by different approaches for five SAs used.

**Figure captions**

**Figure S1.** UV-vis absorption spectra of EP-PDI dilute solution in chloroform and polystyrene (PS):EP-PDI (1:1) blend film spin-coated from CB solution.

**Figure S2.** Normalized Raman spectra of PTB7:EP-PDI blend films processed with different SAs.

**Figure S3.** Normalized Raman spectra of F-DTS:EP-PDI blend films processed with different SAs.

**Figure S4.** (a) UV-vis absorption spectra of two F-DTS:EP-PDI (1:2) blend films with 0.75% CN. (b) J-V curves of organic solar cells based on F-DTS:EP-PDI (1:2, w/w) with 0.75% CN. One blend film was spin-coated at 1500 rpm for 30 seconds (black line). Another blend film was spin-coated at 1500 rpm for 10 seconds and was transferred to a sealed container immediately (red line).

Table S1.

Solvent	Boiling point [°C]	Solubility parameter[MPa <sup>1/2</sup> ]		Solubility [mg mL <sup>-1</sup> ]		
		Small and Hoy <sup>a)</sup>	Fedors <sup>b)</sup>	PTB7	F-DTS	EP-PDI
DIO	333	19.7	20.1	< 0.01	< 1	< 1
ODT	270	18.8	19.2	< 0.01	< 1	< 1
CN	259	21.9	24.4	> 50	> 50	> 100
BT	221	21.8	25.9	> 1	> 20	> 50
BF	173	22.9	24.4	< 0.01	< 0.01	> 50

<sup>a)</sup> Calculated values using Small and Hoy's group contribution theory. <sup>b)</sup> Calculated values using Fedors's group contribution theory.

Figure S1.

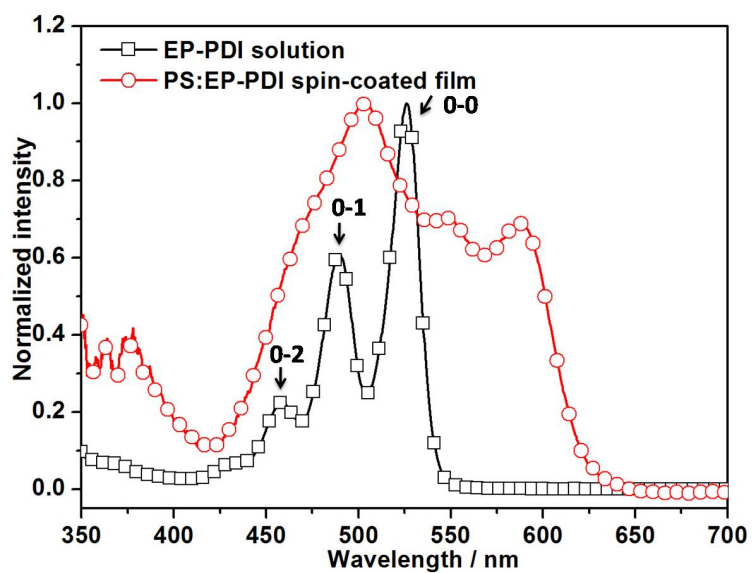


Figure S2.

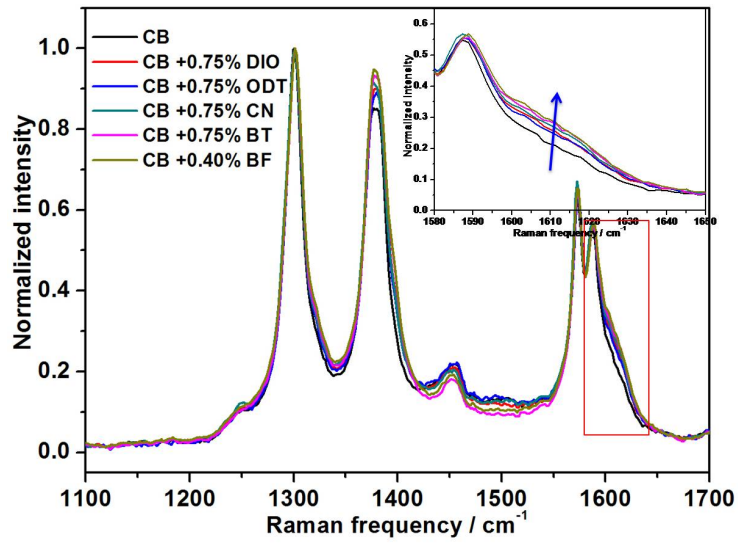


Figure S3.

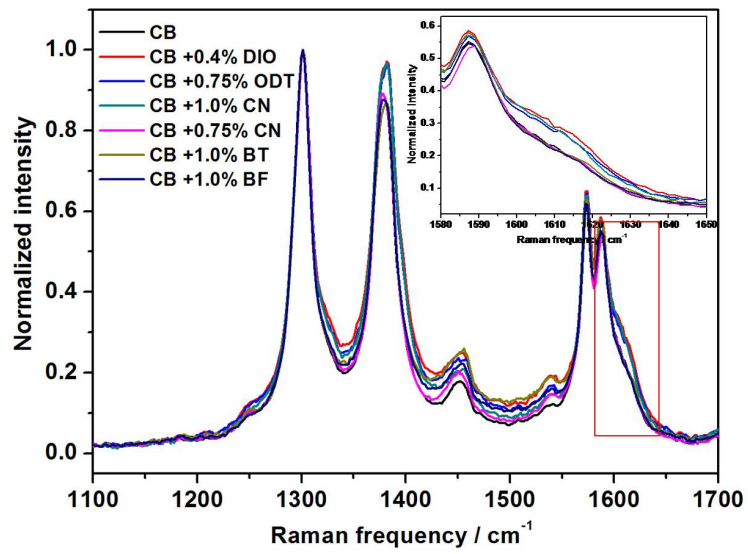
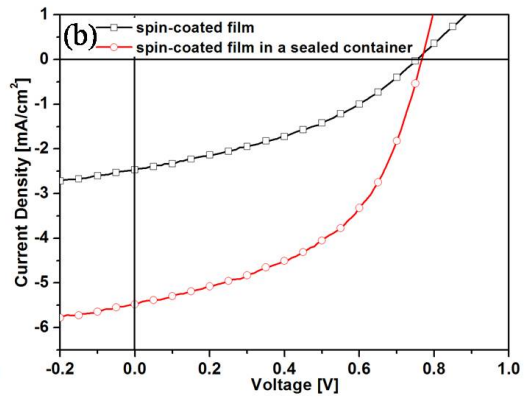
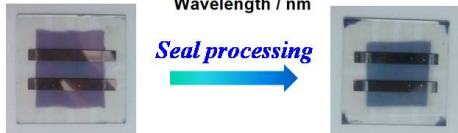
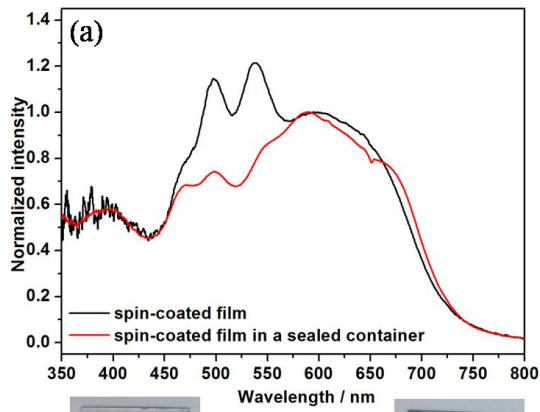


Figure S4.



	<b>Voc</b> <b>(V)</b>	<b>Jsc</b> <b>(<math>\text{mA}/\text{cm}^2</math>)</b>	<b>FF</b>	<b>PCE</b> <b>(%)</b>
Pristine film	0.75	2.469	0.39	0.727
Seal processing	0.77	5.480	0.49	2.074